

Trion dynamics in coupled double quantum wells. Electron density effects.

P. Aceituno* and A. Hernández-Cabrera

Departamento de Física Básica, Universidad de La Laguna, La Laguna 38206-Tenerife, Spain.

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We have studied the coherent dynamics of injected electrons when they are either free or bounded both in excitons and in trions (charged excitons). We have considered a remotely doped asymmetric double quantum well where an excess of free electrons and the direct created excitons generate trions. We have used the matrix density formalism to analyze the electron dynamics for different concentration of the three species. Calculations show a significant modification of the free electron inter-sublevel oscillations cWe have studied the coherent dynamics of injected electrons when they are aused by electrons bound in excitons and trions. Based on the present calculations we propose a method to detect trions through the emitted electromagnetic radiation or the current density.

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I. INTRODUCTION

Coherent oscillations (quantum beats) of the electronic charge in double quantum wells (DQW's) have been widely studied both experimentally and theoretically^{1,2,3,4,5,6,7,8}. The most usual method to achieve charge generation in these structures is the photoexcitation by an ultrashort laser pulse, which causes the same concentration of photoexcited electrons and holes and so, the corresponding excitons. Another possibility consists on injecting electrons and holes by doping two regions close to the structure^{9,10}. Injection offers the advantage of preventing the interaction between the electromagnetic field associated to the excitation and the excited electrons. Here excitons are directly created by hole-assisted electron resonant tunneling (see Fig. 1). Another characteristic of this type of excitons (the so-called direct-created excitons) is that they have spatial coherence, with an in-plane momentum $\mathbf{k}_{exc||} \sim \mathbf{0}$, and can radiate in the direction perpendicular to the DQW. Thus, direct-created excitons are good candidates for their application in vertical planar microcavities. Moreover, injection has two additional advantages: the control of the electron and hole densities by means of the n (donor) or p (acceptor) impurity concentration plus an external applied electric field¹¹, and the avoidance of exciton thermalization. This thermalization is an inevitable consequence of non-tuned laser excitation.

Recently, trions (charged excitons) have been observed in photoexcited doped semiconductor quantum wells. The doping is necessary to get an excess of electrons. These electrons together with the excitons lead to the generation of the negatively charged excitons^{11,12,13}. These bound complexes of three particles have a binding energy large enough to make them observable. The experimental technique most widely used to study trions is time resolved photoluminescence^{14,15}. Processes as generation and recombination of excitons and trions have been recently analyzed through this technique¹⁶.

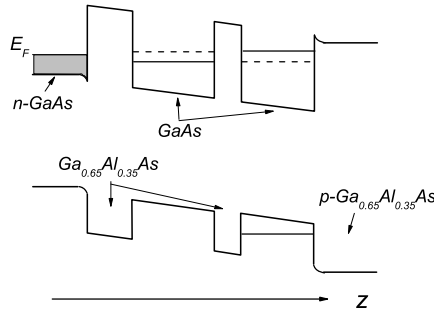


FIG. 1: Scheme of the asymmetric double quantum well structure under free electron resonant condition.

In this work we propose a new method to generate trions in coupled DQW through the direct-created excitons and the necessary free electron excess. In the usual experimental conditions, donor concentration is greater than acceptor concentration ($\sim 10^{18}$ and $\sim 10^{16}$ cm^{-3} , respectively). The process of trion generation can be considered in three steps: first, if the Fermi level of the n -type material (see Fig. 1) resonates with the electronic level in the left-hand wide quantum well (LQW), electrons tunnel to LQW forming the cross or spatially indirect (interwell) excitons with holes diffused into the narrow right-hand well (RQW). Second, if the resonance condition for electrons in the excitonic state between both wells is satisfied, electrons in the excitonic state tunnel to the RQW forming spatially direct (intrawell) excitons. Third, if there is an excess of electrons with respect to holes, free electrons will coexist together with direct-created excitons. Some of these free electrons can be bound to excitons leading to trions. It should be noted that, in coupled quantum wells, the strong dipole-dipole repulsion between excitons prevents the formation of biexcitons. Thus, we will neglect the contributions from these neutral species¹⁷.

Under resonance conditions the charge density in coupled DQWs oscillates between both wells leading to an oscillating dipole moment and the corresponding Terahertz radiation emission. If we consider the holes confined in the well where they were initially injected (due to their large effective masses), the electronic charge will play the main role in the coherent process. Because we will deal with three different types of electron binding (free electron, excitonic electron and trionic electron) they will occupy different energy levels. Moreover, the resonance conditions will be slightly different for each case and thus, their oscillatory behavior will be different as well.

The aim of this paper is the theoretical analysis of the electron in trion dynamics in presence of direct-created excitons and free electrons. We will carry out this in the framework of the matrix density formalism, taking into account the different generation, recombination and annihilation rates for electrons with different type of binding. The dynamics of coexisting excitons and free electrons was studied elsewhere¹⁰. The work is organized as follows: in Sec. I we present the exciton and trion Hamiltonians and the corresponding Schrödinger equations, as well as the dynamics of the electronic density evolution. In Sec. II we include numerical results for the charge density dynamics and for the current density. We also include some comments regarding the results and the used approximations.

II. FORMALISM

Initially, a group of electrons tunnels into the LQW. The charge, dynamically trapped within the wells, produces a reaction field that modifies the structure profile and then, the time evolution of the system. Thus, we need to calculate wave functions for the free-electron, the excitonic electron and the trionic electron, and the corresponding electronic levels, by solving self-consistently the one-particle Schrödinger equation, together with the Poisson equation for each species.

Several techniques have been used to solve the Schrödinger equation for neutral excitons and charged excitons (trions). We can cite the *commutation technique*, developed by Combescot et al.¹⁸. Although it is a very attractive formalism, it is almost inapplicable from the numerical point of view. Another technique, based on the deformed correlated Gaussian wave functions, is the *Stochastic variational method* used by Riva et al.¹⁹. Whittaker and Shields²⁰ used a set of *Landau-level basis* to study the in-plane (xy) motion. These two methods are specially indicated when an external magnetic field is applied. In presence of longitudinal electric fields and in absence of magnetic fields Dacal and Brum²¹ have developed a technique based on the *configuration interaction method (CIM)*²². Because of its feasibility we will use this last technique with some implementations.

The *CIM* considers that the longitudinal (z) and transversal (xy) parts of the wave functions are separable. Thus, we first factorize wave functions of excitonic and trionic electrons into the corresponding in-plane part and confinement (growth) direction part. We can't neglect the in-plane transversal motion part of the Hamiltonian because both parts of the split Hamiltonian are connected by the Coulomb potential. Such a circumstance affects wave functions of the different types of electrons. In order to reach the resonance for the different electronic states we have included in calculations an external electric field applied in the growth direction. The confinement part of the Hamiltonian can be solved by means of the Airy functions within the effective mass approach for electrons and holes. To solve the in-plane part of the one particle Hamiltonian we will follow the same steps as in Ref.²¹. First we will use polar coordinates in

terms of the center of mass (CM) and relative to the hole coordinates. To say,

$$\begin{aligned}\rho_1 &= \rho_{e1} - \rho_h, \\ \rho_2 &= \rho_{e2} - \rho_h, \\ \rho_{CM} &= \frac{\rho_{e1} + \rho_{e2} + m_{h\perp}\rho_h}{m_{h\perp} + 2m_e}.\end{aligned}\tag{1}$$

where $m_{h\perp}$ is the in-plane hole effective mass. We have considered an isotropic electron effective mass.

A. Exciton and trion Hamiltonians

The excitonic Hamiltonian can be written as

$$H_{exc} = H(z_e) + H(z_h) + H_\perp + V_c,\tag{2}$$

where

$$H(z_{e,h}) = -\frac{\hbar^2 \partial}{2\partial z_{e,h}} \frac{1}{m_{e,hz}} \frac{\partial}{\partial z_{e,h}} + V_{DQW}(z_{e,h}) + V_H(z_{e,h}) \pm |e| F z_{e,h}.\tag{3}$$

Here m_{hz} is the hole effective mass in the growth direction, $V_{DQW}(z_{e,h})$ is the asymmetric DCW potential, F is the electric field required to reach the inter-level resonance. $V_H(z_{e,h})$ is the Hartree potential caused by the Coulomb interaction of the electrons with themselves and with the donors (or holes with acceptors). The self-consistent potential $V_H(z_{e,h})$ can be obtained by solving the Poisson's equation

$$\frac{d^2 V_H(z_{e,h})}{dz_{e,h}^2} = \frac{4\pi e^2}{\epsilon} \left[N_{D,A}(z_{e,h}) - \sum_j n_j |\phi_j(z_{e,h})|^2 \right],\tag{4}$$

where $N_{D,A}$ is the doping profile; thus, $\int N_{D,A}(z_e) dz_{e,h} = \sum_j n_{je,h} = n_{2De,h}$ is the in-plane averaged $2De,h$ density of electrons (donors) or holes (acceptors) and j refers to the occupied subbands.

The in-plane term reads

$$H_\perp = -\frac{\hbar^2}{2\mu} \left[\frac{1}{\rho} \frac{\partial}{\partial \rho} \left(\rho \frac{\partial}{\partial \rho} \right) + \frac{1}{\rho^2} \frac{\partial^2}{\partial \rho^2} \right],\tag{5}$$

where we have considered invariant the in-plane excitonic μ reduced mass. The Coulomb potential is

$$V_{ce,h}(\rho) = \int \int dz_e dz_{e',h} \phi_e^2(z_e) \phi_{e',h}^2(z_{e',h}) \frac{e^2}{\epsilon \sqrt{\rho^2 + \langle z_e - z_{e',h} \rangle^2}},\tag{6}$$

and includes electron-hole ($e-h$) and electron-electron ($e-e'$) interactions. This Coulomb effective potential is valid for excitons and for trions, being slightly different for each case due to their different electron wave functions and expected values of $\langle z - z' \rangle$. We will assume a constant dielectric function ϵ across the interfaces. For practical reasons we change the notation of the above Coulomb potential to

$$V_{ij}(\rho) = \frac{e_i e_j}{\epsilon} \int \int dz dz' \frac{\phi_i^2(z) \phi_j^2(z')}{\sqrt{\rho^2 + \langle z - z' \rangle^2}},\tag{7}$$

where $e_i = -e$ for $i = u, l$ and $e_j = e$ ($-e$) for $j = h$ (u, l). Indexes u, l, h refer to electrons in the upper or lower levels and to holes in the right-hand QW, respectively.

The trionic Hamiltonian can be written in an analogous way to the excitonic case by using relative coordinates for the transversal motion:

$$H_{tr} = \sum_{i=1,2} [H(z_{ei}) + H_{\perp i}] + H(z_h) + \frac{1}{m_{h\perp}} \mathbf{p}_1 \mathbf{p}_2 + \frac{e^2}{\epsilon} \frac{1}{\sqrt{|\boldsymbol{\rho}_1 - \boldsymbol{\rho}_2|^2 + (z_{e1} - z_{e2})^2}} \quad (8)$$

where \mathbf{p}_i is the in-plane linear momentum for the i th particle,

$$H(z_{ei,h}) = -\frac{\hbar^2}{2} \frac{\partial}{\partial z_{ei,h}} \frac{1}{m_{ei,hz}} \frac{\partial}{\partial z_{ei,h}} + V_{DQW}(z_{ei,h}) \pm |e| F z_{ei,h}, \quad (9)$$

and

$$H_{\perp i} = -\frac{\hbar^2}{2\mu} \left[\frac{1}{\rho_i} \frac{\partial}{\partial \rho_i} \left(\rho_i \frac{\partial}{\partial \rho_i} \right) + \frac{1}{\rho_i^2} \frac{\partial^2}{\partial \rho_i^2} \right] - \frac{e^2}{\epsilon \sqrt{(z_{ei} - z_h)^2 + \rho_i^2}}. \quad (10)$$

To solve the above Hamiltonians we will follow the same steps as Ref.²¹. Thus, through the *CIM* we construct a non-orthogonal basis set of Slater determinants to solve the eigenvalue problem. This method is widely described in Ref.²². We build up the exciton trial wave function as

$$\Psi_{exc}(z_e, z_h, \boldsymbol{\rho}) = \sum_{ijk} c_{ijk} N_{ijk} \phi(z_e) \phi(z_h) \varphi_k^m(\boldsymbol{\rho}), \quad (11)$$

where c_{ijk} is a variational parameter, N_{ijk} is the determinant normalization, indexes ijk refers to one particle i , j , k , respectively, $\boldsymbol{\rho} = \boldsymbol{\rho}_e - \boldsymbol{\rho}_h$, m is the relative particle angular momentum, and $\phi(z_{e,h})$ is the envelope wave function, and $\varphi_k^m(\boldsymbol{\rho})$ is the one particle in-plane wave function

$$\varphi_k^m(\boldsymbol{\rho}) = N_{km} \rho^m \exp \left[-\frac{\rho^2}{\lambda_k^2} \right] \exp(im\theta). \quad (12)$$

Here N_{km} is the relative particle normalization factor and λ_k is a set of parameters (one for each angular momentum m) that we have taken from Ref.²³.

The above expressions can be generalized for trions as

$$\begin{aligned} \Psi_{tr}(z_{e1}, z_{e2}, z_h, \boldsymbol{\rho}_1, \boldsymbol{\rho}_2) &= \sum_{ijmnpqr} c_{ijmnpqr} N_{ijmnpqr} \phi_p(z_h) \\ &\times \left[\begin{aligned} &\phi_q(z_{e1}) \varphi_i^m(\boldsymbol{\rho}_1) \phi_r(z_{e2}) \varphi_j^n(\boldsymbol{\rho}_2) \\ &+ \phi_r(z_{e1}) \varphi_j^n(\boldsymbol{\rho}_1) \phi_q(z_{e2}) \varphi_i^m(\boldsymbol{\rho}_2) \end{aligned} \right]. \end{aligned} \quad (13)$$

It is important to remark that, in absence of external magnetic fields, the only bound state for trions is the singlet $m + n = 0$. We would like to mention here that we have included a new contribution: the spatial bending of the structure due to the doping. Thus, after variationally obtaining the above wave functions for $V_H(z_{e,h}) = 0$, we need to solve the Poisson equations (4) and turn again to the Slater Hamiltonians. Fortunately, we only need to modify one particle envelope functions in the growth direction by solving self-consistently the z -direction part of the corresponding Schrödinger equations (3, 9) together with the Poisson equations. For practical purposes, to numerically work the z -direction part with the Transfer Matrix Method is better than using the Airy functions. Variational coefficients and normalization functions for wave functions (11, 13) will be slightly different when including the spacial self-consistent potential $V_H(z_{e,h})$. Based on Ref.²² we use λ_k parameters for the in-plane part of the Hamiltonians corresponding to s and p^\pm states. The inclusion of the d^\pm states only affects about a 5% the position of the excitonic and trionic energy levels but it supposes an additional computational effort which is not reflected on the final results. Another key point consists on the identification of the levels because it will be fundamental in the next section. This information is contained in the envelope functions $\phi(z_{e,h})$. Thus, both for electrons and holes, the two deepest levels correspond to

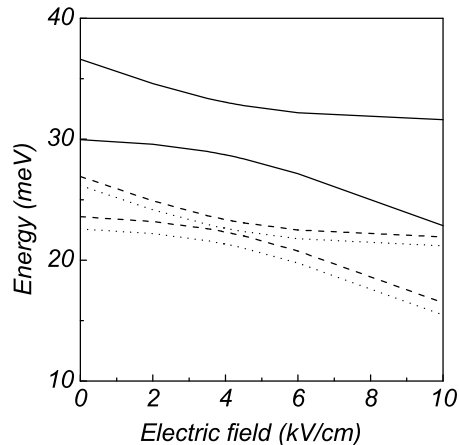


FIG. 2: Energy levels versus applied electric field. Solid line: free electrons. Dashed line: excitonic electrons. Dotted line: trionic electrons.

the resonant levels of the two coupled wells. Hereinafter we will distinguish them with the indexes u (upper) and l (lower). We have neglected the possible hole states mixing for the applied electric fields under consideration because we are mainly interested in the conduction band states.

Concerning to free-electron, the electronic levels (29.97 meV and 36.61 meV) would correspond to the LQW and the RQW, respectively, if they were decoupled. For the excitonic electron case these two levels are shifted by the Coulomb interaction by different energy amounts corresponding to the indirect and direct exciton¹⁰ (5.78 meV and 8.60 meV , respectively). We have considered that hole energy levels remain the same due to the bigger hole effective mass. The case of trions is more complicated due to two possible additional forms of setting the new electron with regard to the previous excitonic state. From the energy point of view, the most favorable cases correspond to the direct exciton bounded to an electron either in the LQW or in the RQW. That means a shift of the free electron levels of 6.91 meV and 9.93 meV , respectively. We represent in Fig. 2 electronic levels as function of the external electric field. It can be seen that the resonances of the three species take place for almost the same applied electric field. With these values we calculate the level splitting for coupled wells, Δ_T , and the transmission matrix element, T , which equals to $\Delta_T/2$ at resonance. The level splitting energy for decoupled wells is $\Delta = \sqrt{\Delta_T^2 - 4T^2}$, for each case. We will use these parameters in the next subsection.

B. Time evolution of the electron density

In this Section we will treat the dynamic behavior of the electronic density within the Matrix Density formalism in the momentum representation. Although electrons spend some time in tunneling the left-hand confining barrier, we assume an ultrafast $\delta(t)$ injection of electrons in the LQW to simplify calculations. Also, diffusion of holes from the p -doped material to the RQW is considered as a $\delta(t)$ function. We use this approximation because we are mainly interested in the dynamics of the electronic density between wells after injection. Based on the previous premises, we start from the general quantum kinetic equation for the density operator $\hat{R}(t)$

$$\frac{\partial \hat{R}(t)}{\partial t} + \frac{i}{\hbar} [\hat{H}, \hat{R}(t)] = 0. \quad (14)$$

Here \hat{H} is the DQW many-band Hamiltonian we have described above. If we project on the conduction band states we find the 2×2 matrix kinetic equation in the momentum representation

$$\begin{aligned} & \frac{\partial [\hat{\rho}_{\mathbf{p}}(t)]_{jj'}}{\partial t} + \frac{i}{\hbar} [\hat{h}, \hat{\rho}_{\mathbf{p}}(t)]_{jj'} - \frac{i}{\hbar} \langle [V_c, a_{j's\mathbf{p}}^\dagger a_{js\mathbf{p}}] \rangle \\ &= [\hat{G}_{\mathbf{p}}(t)]_{jj'}. \end{aligned} \quad (15)$$

This equation describes the time evolution of the density matrix $\hat{\rho}_{\mathbf{p}}(t)$, where indexes j, j' refer to the levels u (upper) and l (lower) of the DQW, and \mathbf{p} is the electronic wave vector. In the expression (15) $[\hat{\rho}_{\mathbf{p}}(t)]_{jj'} = \langle a_{j's\mathbf{p}}^\dagger a_{js\mathbf{p}} \rangle$, where $a_{j's\mathbf{p}}^\dagger$ and $a_{js\mathbf{p}}$ are the creation and annihilation electron operators, s is the electronic spin, and $\langle \rangle$ means the statistical average. The operator $\hat{h} = (\Delta/2)\hat{\sigma}_z + T\hat{\sigma}_x$ is the matrix of the one-electron DQW Hamiltonian without Coulomb interactions, and $\hat{\sigma}_i$ ($i = x, y, z$) are the standard Pauli matrices. For numerical and analytical reasons it is convenient to remove the Coulomb interaction from Hamiltonians (3) and (10) and to work it separately in the Hartree representation. Thus, variational coefficients and normalization factors in $\phi_k^m(\boldsymbol{\rho})$ (13) will be slightly different. The Coulomb interaction can be written as

$$V_c = \frac{1}{2S} \sum_{ij} \sum_{ss'} \sum_{\mathbf{k}\mathbf{k}'\mathbf{q}} M_{ij}(q) a_{is\mathbf{k}+\mathbf{q}}^\dagger a_{js'\mathbf{k}'-\mathbf{q}}^\dagger a_{js'\mathbf{k}'} a_{is\mathbf{k}}, \quad (16)$$

where i, j refer both to electron and hole states to include electron-electron, electron-hole and hole-hole Coulomb interactions. S is the normalization area and $q = |\mathbf{q}|$. The matrix elements $M_{ij}(q)$ are defined as²³

$$M_{ij}(q) = -\frac{2\pi e_i e_j}{\epsilon q} \iint dz dz' \exp(-q|z - z'|) |\phi_i(z)|^2 |\phi_j(z')|^2, \quad (17)$$

where $e_{i(j)} = -e$ for electrons $e_{i(j)} = e$ for holes (e is the electron charge).

After substitution of (16) in (15) we reach

$$\frac{\partial \hat{\rho}_{\mathbf{p}}(t)}{\partial t} + \frac{i}{\hbar} [\hat{h} + \hat{Q}(t) + \hat{X}_{\mathbf{p}}(t), \hat{\rho}_{\mathbf{p}}(t)] = \hat{G}_{\mathbf{p}}(t), \quad (18)$$

where $\hat{Q}(t)$ is the direct Coulomb potential and $\hat{X}_{\mathbf{p}}(t)$ is the exchange potential. If we define the projectors over the l and u conduction states as $\hat{P}_l = (1 + \hat{\sigma}_z)$ and $\hat{P}_u = (1 - \hat{\sigma}_z)$, respectively, $n_h(t)$ is the hole concentration in the h -th hole subband, and $n_{l,u}(t) = (2/S) \sum_{\mathbf{p}} \text{Tr} [\hat{P}_{l,u}(t) \hat{\rho}(t)]$ is the electron concentration in the upper (u) and lower (l) levels, thus matrices $\hat{Q}(t)$ and $\hat{X}_{\mathbf{p}}(t)$ can be written as

$$\begin{aligned} \hat{Q}(t) &= \left[M_{ll}(q)n_l(t) + M_{lu}(q)n_u(t) - \sum_h M_{lh}(q)n_h(t) \right]_{q \rightarrow 0} \hat{P}_l \\ &\quad - \left[M_{uu}(q)n_u(t) + M_{ul}(q)n_l(t) - \sum_h M_{uh}(q)n_h(t) \right]_{q \rightarrow 0} \hat{P}_u, \end{aligned} \quad (19)$$

$$\hat{X}_{\mathbf{p}}(t) = \frac{1}{S} \sum_{\mathbf{q}} \left\{ \begin{aligned} & [M_{ll}(q) - M_{lu}(q)] \text{Tr} \left(\hat{P}_l \hat{\rho}_{\mathbf{p}-\mathbf{q}}(t) \right) \hat{P}_l \\ & + [M_{uu}(q) - M_{lu}(q)] \text{Tr} \left(\hat{P}_u \hat{\rho}_{\mathbf{p}-\mathbf{q}}(t) \right) \hat{P}_u \\ & + M_{lu}(q) \hat{\rho}_{\mathbf{p}-\mathbf{q}}(t) \end{aligned} \right\}. \quad (20)$$

After integrating (18) over momentum \mathbf{p} one finds that the non-diagonal part of the exchange potential, $M_{lu}(q) \hat{\rho}_{\mathbf{p}-\mathbf{q}}(t)$, does not contribute to the commutator in eq. (18). The diagonal part of $\hat{X}_{\mathbf{p}}(t)$, together with

the Coulomb potential $\widehat{Q}(t)$, provides us the renormalization of the levels and, thus, the renormalization of the splitting Δ because these local terms couple different subbands. It is important to point out that, contrary to Ref. (3), in our case we are dealing with typical values of q of the order of 10^8 m^{-1} , which is the same order as the distance between the centers of the wells Z . Thus, we cannot neglect the exchange contribution.

Now, we define the concentration matrix as $\widehat{n}(t) = (2/S) \sum_{\mathbf{p}} \widehat{\rho}_{\mathbf{p}}(t)$ and integrate over \mathbf{p} to obtain the isospin representation^{2,3}:

$$\widehat{n}(t) = n_0(t) + \sum_i n_i(t) \widehat{\sigma}_i(t) = n_0(t) + \mathbf{n}(t) \widehat{\boldsymbol{\sigma}}. \quad (21)$$

After substituting in (18) and introducing the subindex k to differ free electron (f), excitonic electron (exc) and trionic electron (tr), the above Liouville (18) equation leads to the Bloch-equation system for the evolution of the isospin density vector $\mathbf{n}_k(t) = (n_k^x(t), n_k^y(t), n_k^z(t))$ and the scalar density $n_k^0(t)$:

$$\begin{aligned} \frac{\partial \mathbf{n}_k(t)}{\partial t} - [\mathbf{L}_k(t) \times \mathbf{n}_k(t)] &= \mathbf{G}_k(t) - \mathbf{\Lambda}_k(t), \\ \frac{\partial n_k^0(t)}{\partial t} &= N_k \delta(t) - S_k(t). \end{aligned} \quad (22)$$

where $N_k \delta(t) - S_k(t) = (1/S) \sum_{\mathbf{p}} Tr(\widehat{G}_{\mathbf{p}k}(t))$ and $\mathbf{G}_k(t) - \mathbf{\Lambda}_k(t) = (1/S) \sum_{\mathbf{p}} Tr(\widehat{\boldsymbol{\sigma}} \widehat{G}_{\mathbf{p}k}(t))$. The nonlinear dynamic properties of the system are described by $\mathbf{L}_k(t) = (2T_k/\hbar, 0, \Delta_k(t)/\hbar)$, where $\Delta_k(t)$ is the renormalized time dependent level splitting induced by the carrier density and T_k is the tunneling matrix element. $\mathbf{G}_k(t) = (0, 0, N_k \delta(t))$ is the generation term and N_k denotes the initial density of electrons. In the present case, just at $t = 0$, $N_f = N_e$ (injected electron density) and $N_{exc, tr} = 0$. The terms $\mathbf{\Lambda}_k(t)$ and $S_k(t)$ include generation and annihilation rates for excitons and trions. We will describe them below. Vector equations (22) split into three coupled Bloch system, corresponding to free electrons, excitonic electrons and trionic electrons. The time dependent energy splitting renormalization is determined (after an easy but tedious algebra) by the many-body Coulomb interaction through the Hartre-Fock approximation (19,20)

$$\begin{aligned} \Delta_{exc(tr)}(t) &= \Delta_{exc(tr)} + \frac{2\pi e^2}{\epsilon} \int \int dz dz' |z - z'| \\ &\times \left[\left| \phi_{u,exc(tr)}(z') \right|^2 - \left| \phi_{l,exc(tr)}(z') \right|^2 \right] \\ &\times \left[\left(\left| \phi_{u,exc(tr)}(z) \right|^2 n_{exc(tr)}^u(t) + \left| \phi_{l,exc(tr)}(z) \right|^2 n_{exc(tr)}^l(t) \right) \right. \\ &\quad \left. - \sum_h \left| \phi_h(z) \right|^2 n^h(t) \right] \\ &- \alpha \frac{2\pi e^2}{\epsilon} \int \int dz dz' \delta(z - z') \left[\left| \phi_{l,exc(tr)}(z') \right|^2 - \left| \phi_{u,exc(tr)}(z') \right|^2 \right] \\ &\times \left[\left| \phi_{l,exc(tr)}(z) \right|^2 n_{exc(tr)}^l(t) \right. \\ &\quad \left. - \left| \phi_{u,exc(tr)}(z) \right|^2 n_{exc(tr)}^u(t) \right], \end{aligned} \quad (23)$$

and

$$\begin{aligned}
\Delta_f(t) = & \Delta_f + \frac{2\pi e^2}{\epsilon} \int \int dz dz' |z - z'| \\
& \times \left[|\phi_{u,f}(z')|^2 - |\phi_{l,f}(z')|^2 \right] \\
& \times \left[|\phi_{u,f}(z)|^2 n_f^u(t) + |\phi_{l,f}(z)|^2 n_f^l(t) \right] \\
& - \alpha \frac{2\pi e^2}{\epsilon} \int \int dz dz' \delta(z - z') \left[|\phi_{l,f}(z')|^2 - |\phi_{u,f}(z')|^2 \right] \\
& \times \left[\begin{array}{c} |\phi_{l,f}(z)|^2 n_f^l(t) \\ - |\phi_{u,f}(z)|^2 n_f^u(t) \end{array} \right], \tag{24}
\end{aligned}$$

where $n_k^{u(l)}(t)$ and $n_h(t)$ are the electronic density in the upper (lower) level and the hole density in the right-hand well, respectively. The last terms in (23) and (24), proportional to the coefficient α , correspond to the exchange interaction contribution. To calculate this contribution we need to introduce the carrier screening since the use of the bare exchange potential may lead to divergent results due to its singularity when $\mathbf{q} \rightarrow \mathbf{0}$. This means that the energy renormalization caused by the exchange term may diverge. In an easy approach, and considering parabolic subbands filled by degenerate electrons, we use the statically screened Coulomb potential $V_s(\mathbf{q}) = \frac{4\pi e^2}{\epsilon(q+q_s)}$, where $q_s = \frac{2me^2}{\epsilon\hbar^2}$ is the screening wavevector. In this case, the screening exchange can be written as $\sum_{\mathbf{q}} V_s(\mathbf{q}) [\hat{\rho}(\mathbf{p} - \mathbf{q})]_{jj} + \frac{1}{2} \lim_{r \rightarrow 0} [V_s(r) - V(r)]$.

The last term corresponds to the difference between the screened and unscreened Coulomb potential, in the real space, at $r = 0$. Integration over \mathbf{p} and \mathbf{q} of the exchange commutator in (18) leads to corrections of the order of 11% in the level splitting. In the present work $\alpha \approx 12.1\text{\AA}$. Exact calculations lead to three different values of the parameter α , but differences (of about 10%) are not visible in results.

We will assume that only the first heavy hole level is occupied. It is worth mentioning that excitonic and trionic electrons not only interact among themselves and with holes but with free electrons as well. It is important to remember that, for direct-created excitons in DQWs, holes and electrons are generated in different wells. Equations (23, 24) differ from similar expression in Ref.³ due to the exchange contribution.

If we denote by N_h as the initial total hole density, then $n_h(t=0) = N_h$ and

$$n_h(t) = n_{exc}^u(t) + n_{exc}^l(t) + \frac{n_{tr}^u(t) + n_{tr}^l(t)}{2}. \tag{25}$$

. The factor 1/2 in the last addendum appears because n_{tr}^j denotes the trionic electron density which is twice the hole density forming trions. The scalar density $n_k^0(t)$ is the sheet electron density (the total density of electrons in the DQW structure). Also, we can write

$$\begin{aligned}
n_k^0(t) &= n_k^l + n_k^u(t), \\
n_k^z(t) &= n_k^l(t) - n_k^u(t). \tag{26}
\end{aligned}$$

Now we define τ as the relaxation time for the non-diagonal part of the density matrix^{3,25}. This relaxation leads to the interlevel redistribution of the electron density. We consider here the generation and annihilation terms. Initially, free electrons are injected quasi-instantaneously and lose their free condition to generate excitons and trions. So we have two annihilation mechanisms expressed through the generation rates Γ_{exc} and Γ_{tr} , respectively. Moreover, trions destruction gives rise to free electrons because the recombination of one electron with the hole leaves an additional free electron. Thus, we have an additional generation term for free electrons at a rate $1/\tau_{tr}$. By writing densities

$n_f^i(t)$ and $n_{exc,tr}^i(t)$ in units of N_e , equation (22) for free electrons becomes

$$\begin{aligned} \frac{d}{dt}n_f^x(t) + \frac{\Delta_f(t)}{\hbar}n_f^y(t) + \frac{1}{\tau}n_f^x(t) &= 0, \\ \frac{d}{dt}n_f^y(t) - \frac{\Delta_f(t)}{\hbar}n_f^x(t) + \frac{2T_f}{\hbar}n_f^z(t) + \frac{1}{\tau}n_f^y(t) &= 0, \\ \frac{d}{dt}n_f^z(t) - \frac{2T_f}{\hbar}n_f^y(t) - \frac{N_f}{N_e}\delta(t) - \frac{1}{\tau_{tr}}n_{tr}^0(t) + \Gamma_{tr}n_m(t) + \Gamma_{exc}n_h(t) &= 0, \\ \frac{d}{dt}n_f^0(t) - \frac{N_f}{N_e}\delta(t) - \frac{1}{\tau_{tr}}n_{tr}^0(t) + \Gamma_{tr}n_m(t) + \Gamma_{exc}n_h(t) &= 0, \end{aligned} \quad (27)$$

where $n_m(t) = \min[n_f^0(t), n_{exc}^0(t)]$. In a similar way excitonic electrons appear, after the instantaneous hole diffusion, at a rate Γ_{exc} and disappear due to trion generation and recombination at rates Γ_{tr} and $1/\tau_{exc}$, respectively. Thus, for excitonic electrons,

$$\begin{aligned} \frac{d}{dt}n_{exc}^x(t) + \frac{\Delta_{exc}(t)}{\hbar}n_{exc}^y(t) + \frac{1}{\tau}n_{exc}^x(t) &= 0, \\ \frac{d}{dt}n_{exc}^y(t) - \frac{\Delta_{exc}(t)}{\hbar}n_{exc}^x(t) + \frac{2T_{exc}}{\hbar}n_{exc}^z(t) + \frac{1}{\tau}n_{exc}^y(t) &= 0, \\ \frac{d}{dt}n_{exc}^z(t) - \frac{2T_{exc}}{\hbar}n_{exc}^y(t) - \Gamma_{exc}n_h(t) + \frac{1}{\tau_{exc}}n_{exc}^0(t) + \Gamma_{tr}n_m(t) &= 0, \\ \frac{d}{dt}n_{exc}^0(t) - \Gamma_{exc}n_h(t) + \frac{1}{\tau_{exc}}n_{exc}^0(t) + \Gamma_{tr}n_m(t) &= 0, \end{aligned} \quad (28)$$

and, for trionic electrons,

$$\begin{aligned} \frac{d}{dt}n_{tr}^x(t) + \frac{\Delta_{tr}(t)}{\hbar}n_{tr}^y(t) + \frac{1}{\tau}n_{tr}^x(t) &= 0, \\ \frac{d}{dt}n_{tr}^y(t) - \frac{\Delta_{tr}(t)}{\hbar}n_{tr}^x(t) + \frac{2T_{tr}}{\hbar}n_{tr}^z(t) + \frac{1}{\tau}n_{tr}^y(t) &= 0, \\ \frac{d}{dt}n_{tr}^z(t) - \frac{2T_{tr}}{\hbar}n_{tr}^y(t) - \Gamma_{tr}n_m(t) + \frac{1}{\tau_{tr}}n_{tr}^0(t) &= 0, \\ \frac{d}{dt}n_{tr}^0(t) - \Gamma_{tr}n_m(t) + \frac{1}{\tau_{tr}}n_{tr}^0(t) &= 0. \end{aligned} \quad (29)$$

In the above expressions Γ_{tr}^{-1} is the mean time required for the trion formation and τ_{exc} and τ_{tr} are the intrinsic exciton and trion lifetimes. An extensive analysis about these times can be found in the paper of Esser et al.¹⁴. We have considered that the process of exciton formation is much faster than the generation of trions and the annihilation of both excitons and trions¹⁶. Actually, Γ_{tr} corresponds to the scattering rate of excitons with free electrons, which gives rise to the trion formation. Experimentally, this scattering formation rate is proportional to the free electron density, to the in-plane exciton area (which contributes to the cross section of the process), and to the kinetic energy of the two interacting species. In our work we have used data for *GaAs* – *GaAlAs* included in¹⁶.

Equations (28), (29), and (30) allow us to get the total density values

$$\begin{aligned} n_T^z(t) &= n_f^z(t) + n_{exc}^z(t) + n_{tr}^z(t), \\ n_T^0(t) &= n_f^0(t) + n_{exc}^0(t) + n_{tr}^0(t), \end{aligned} \quad (30)$$

where $n_T^z(t)$ is proportional to the total dipole moment.

C. The current density

Within the dipole approximation the current density can be written as

$$j_W(t) = \frac{e}{L} \sum_k n_k^z(t) v_{\perp W}^k, \quad (31)$$

where l, l' refer to the coupled levels, $v_{\perp ll'} = -\frac{i\hbar}{m} \langle \phi_l(z) | \frac{\partial}{\partial z} | \phi_{l'}(z) \rangle = v_{\perp lu} = -\frac{i\hbar}{m} \langle \phi_l(z) | \frac{\partial}{\partial z} | \phi_u(z) \rangle$ is the intersubband transverse velocity, and L is the size of the structure. Since we have supposed a left-hand barrier doping to locate the Fermi level just above the two deepest conduction levels, only these two levels will be occupied and we can neglect contributions from remote bands. Therefore, the whole problem is now reduced to the additional calculation of the transverse velocity.

III. RESULTS AND DISCUSSION

We have used in calculations an asymmetric DQW (Fig.1) configured by a 110\AA *GaAs* LQW and a 100\AA *GaAs* RQW, separated by a 30\AA $\text{Ga}_{0.65}\text{Al}_{0.35}\text{As}$ barrier. Then we can guarantee the interaction of the electron wave functions of neighboring wells (non-null overlap). For this structure the distance between the centers of the wells is $Z_c = 135\text{\AA}$. We have also considered the permittivity as constant along the whole structure, $\epsilon = 12.9$. To analyze the contribution of trions to the total dipole moment we perform numerically the coupled system (28-30) through the Runge-Kutta method. We will restrict ourselves to cases in which the density of injected electrons is higher than the density of diffused holes, e.g., an excess of free electron density of about $10^9 - 10^{10} \text{cm}^{-2}$. Higher densities could prevent the trion formation¹².

Figures 3(a-c) represent the density evolution of free, excitonic and trionic electrons $[n_k^0(t)]$ for different N_h/N_e ratios. Initially, there are only free electrons. The density of these electrons decays quickly, giving rise to the excitons. These excitons yield to trions generation due to the scattering processes between excitons and free electrons. The recombination of electron-hole pairs reduces the density of electrons both in exciton and in trion. In the last case each extinct trion leaves one electron free, which contributes to increase the stock of remaining free electrons. That recombination leads to a long time limit of the total density of electrons that equals the free electron excess, $n_T^0(t \rightarrow \infty) = n_f^0(t \rightarrow \infty) = 1 - N_h/N_e$. Cases (a), (b), (c) corresponds to $N_h/N_e = 0.8, 0.5, 0.2$, respectively. At first sight, the formation of trions seems to be more efficient when the initial hole concentration is half the initial free electron concentration. In this case the trionic electron density can even exceed the excitonic electron density.

Next we analyze the behavior of $n_k^z(t)$ to obtain $n_T^z(t)$, which is proportional to the dipole moment. As stated in Eq. (26) $n_k^z(t)$ is the difference between the densities of the lower and the upper energy levels. We represent $n_k^z(t)$ in Panels 4(a-c) for $N_e = 10^{10} \text{cm}^{-2}$. Each figure corresponds to one of the above mentioned N_h/N_e ratios. As expected for low densities, free electron density oscillates between lower and upper levels with an exponential-like envelope profile not only due to the depopulation of the corresponding levels but also to the interlevel redistribution. However, Coulomb interaction forces the excitonic electrons to oscillate staying basically in the upper level before the recombination and the interlevel redistribution. Trionic electron tends to occupy mainly the lower level. On the whole, the mentioned contributions give place to a modulation of the free electron dipole moment which diminishes when N_h/N_e does.

For higher densities the effect of the electron-electron Coulomb interaction becomes more noticeable. Panels 5(a-c) represent $n_k^z(t)$ for $N_e = 5 \times 10^{10} \text{cm}^{-2}$. Free electron density behavior is absolutely different than the low density case and, initially, most of the electrons stay in the lower level and only a few of them oscillates between wells. At the same time excitonic electrons experience a strong confinement in the upper level and trionic electrons do the same in the lower level. The frequency of the three species of electrons shows an increase that is more remarkable for the trion case. As in the low density case, the effect over the free electron dipole moment due to excitons and trions decreases when N_h/N_e decreases. Another effect of the charge density increase is to diminish the oscillation period breaking down the harmonic response of the dipole moment. The anharmonic behavior can be explained as a consequence of the space-charge potential created by the spatial distribution of electrons and holes. This space-charge potential is repulsive for holes and attractive for electrons. For densities higher than the values used in this work ($N_e \gtrsim 10^{11} \text{cm}^{-2}$) the Hartree-Fock potential becomes more important and a better approximation to analyze the carrier distribution would be necessary. However, the Hartree-Fock approximation used here for the many-body Coulomb potential incorporates the main peculiarities of the particle interactions as function of the carrier densities. Thus, electron-hole attraction dominates over electron-electron and hole-hole repulsion at low densities. When the carrier density increases the repulsive part of the Hartree-Fock potential increases as well. Beyond certain initial density of free electrons, the repulsion equals the attractive Coulomb interaction and the oscillation period decreases for the three species. For higher densities the Coulomb interaction of the second electron with the hole in the trion is canceled resulting in the trion extinction, which leads to excitons and free electrons. If the free electron density increases even more, the binding energy of excitons tends to zero and excitons also disappear.

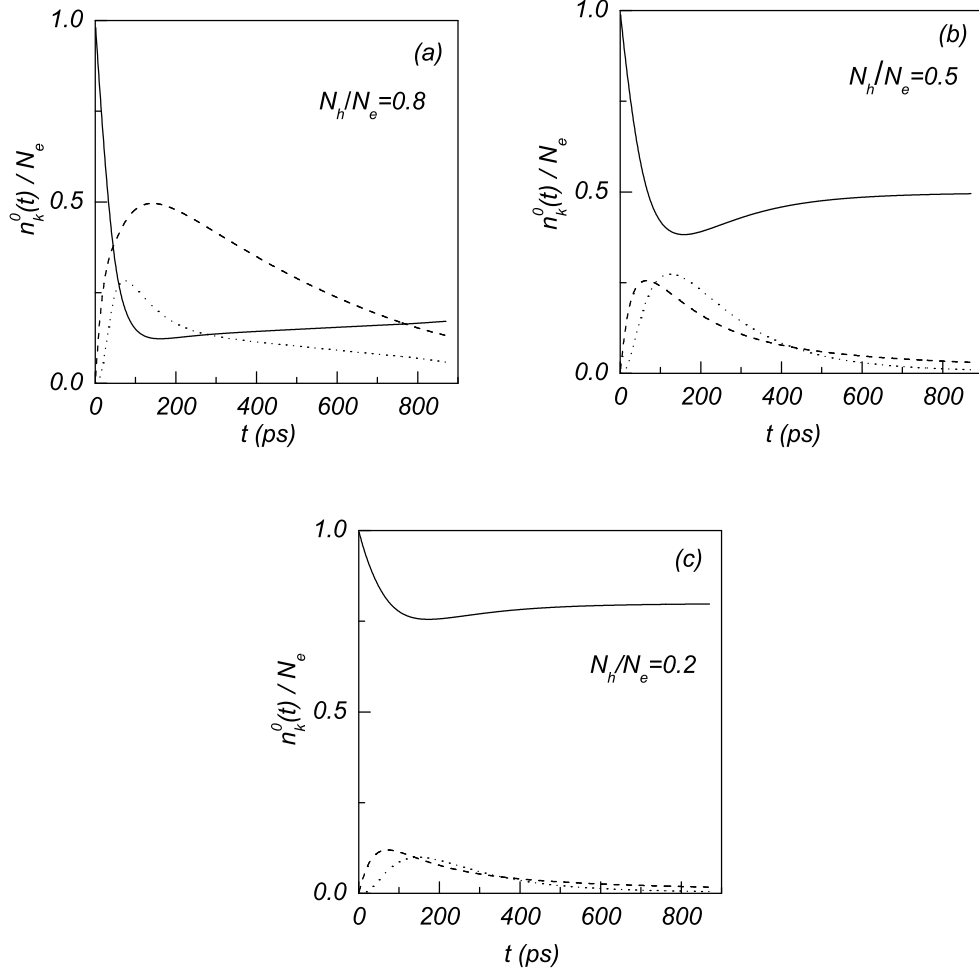


FIG. 3: Temporal evolution of the normalized electron density. (a) $N_h/N_e = 0.8$, (b) $N_h/N_e = 0.5$, and (c) $N_h/N_e = 0.2$. Solid line: free electrons, dashed line: excitons and dotted line: trions.

The shape of the current density is the same as the shape of the $n_T^z(t)$ density and we don't believe necessary to reproduce them here. As an example we include current quantum beats in Fig. 6(a) where upper panel corresponds to the current density for parameters of Fig. 4(a), and lower panel to data of Fig. 5(a). It should be notice that, in any case, current density vanishes for long times due to relaxation as Fig. 6(b) shows for the same parameters as in Fig. 6(a).

Now, we will mention here the approximations used in the present method of calculation. We assumed that the electron injection and hole diffusion are processes much shorter than the coherent oscillation period and the exciton generation time, which, in turn, is even shorter than trion generation and recombination times. Another important point is the possible effect of the applied external electric field on the trions. We have used field intensities lower than $5kV/cm$ to avoid the ionization or the possible trion diffusion along the structure¹³.

In summary, we propose a new method for the generation of trions that avoids the undesirable effects coming from the interaction of the laser electromagnetic field with photoexcited carriers, an ever-present problem in photoexcitation. The method is based on electron tunneling injection and hole diffusion from remotely doped layers⁹. We have also analyzed the effect of these trions, together with the direct-created excitons, on the time dependent dipole moment caused by the free electron density oscillations. Such an effect is only relevant for an initial excess of the free electron density with regard to the hole density. We have studied the above mentioned effect through the three coupled Bloch system obtained from the Liouville equation and taking into account five possible time dependent processes. Our

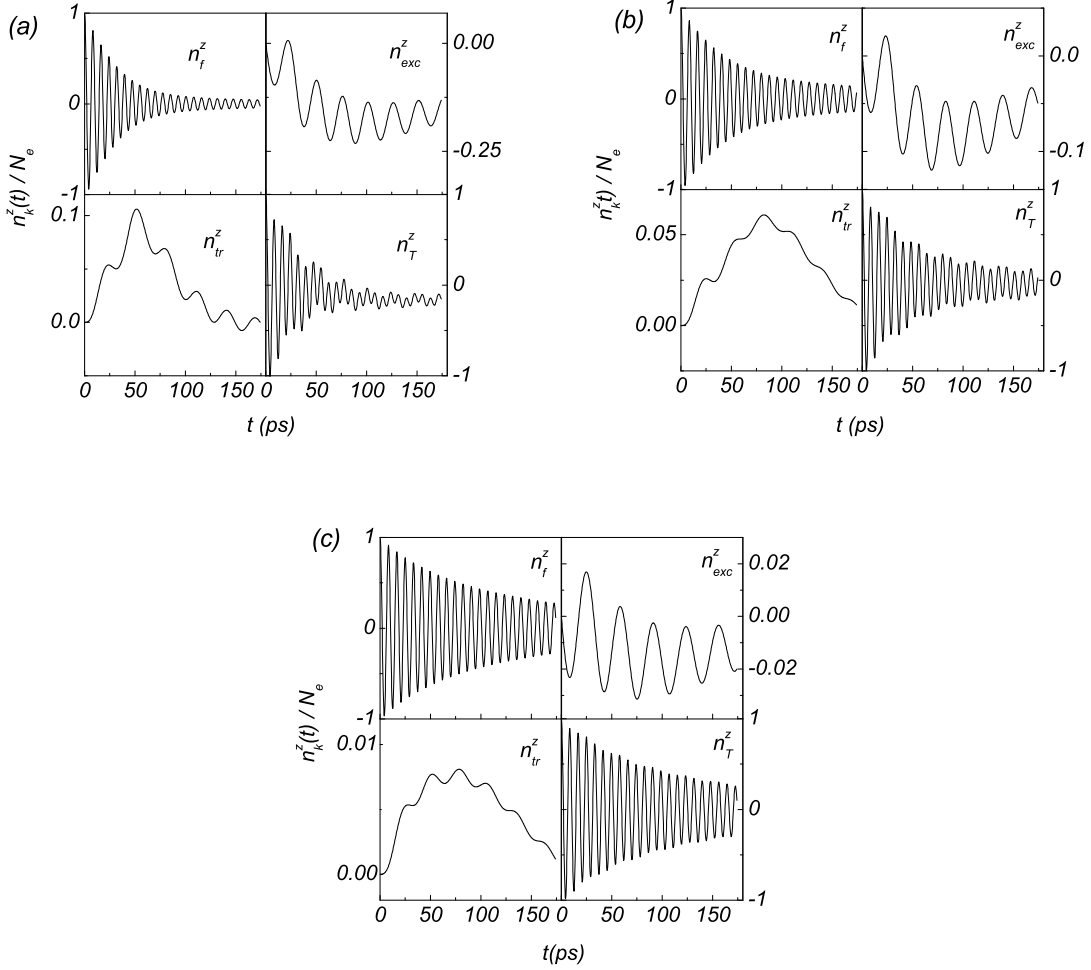


FIG. 4: Temporal evolution of the normalized dipole moment with $N_e = 10^{10} \text{ cm}^{-2}$. Upper panels: free and excitonic electrons. Lower panels: trionic electrons and total dipole moment. (a) $N_h/N_e = 0.8$, (b) $N_h/N_e = 0.5$, and (c) $N_h/N_e = 0.2$.

results show a wide variety of responses, from modulation and quantum beats to anharmonic regime, caused by the different carrier densities of the three types of electrons. These densities mainly affect the dynamics of the level splitting energy of electrons and thus, the coupling strength. The combined electron oscillations yield to a nonlinear coherent electromagnetic radiation from the semiconductor structure. In other words, the existence of trions could be detected by means of the current density, which is straightforwardly translated into Terahertz radiation.

As far as we know only photoluminescence experiments for trions in coupled DQW are available. Moreover, there are not any studies, neither experimental nor theoretical, about the possible effect of trions on the dipolar radiation emission from quantum wells. Therefore, we are forced to extrapolate from well-known results for excitons. Since the standard way to generate trions is ultra-fast photoexcitation, a detailed analysis of this process deserves special attention together with the peculiarities of the high-density case. We expect that the presented work will stimulate experimental studies of the dynamics of direct-created excitons and trions.

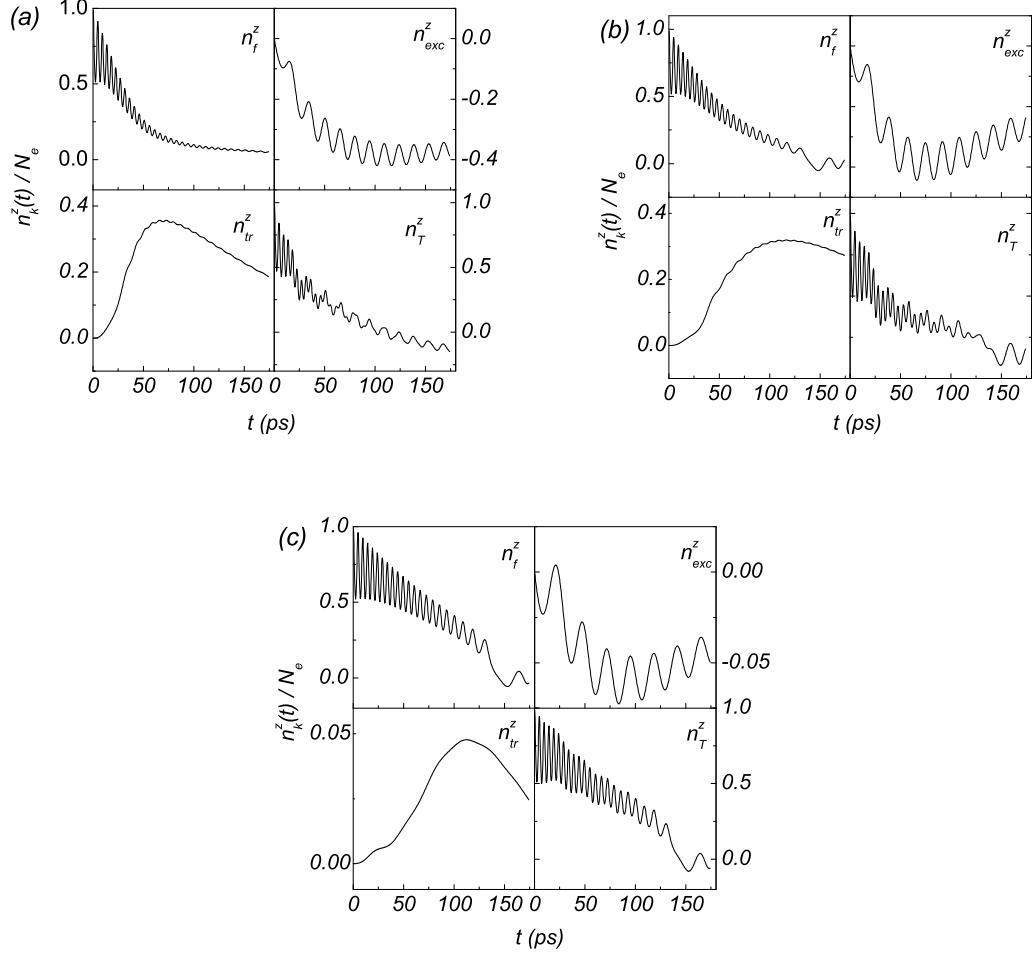


FIG. 5: Temporal evolution of the normalized dipole moment with $N_e = 5 \times 10^{10} \text{ cm}^{-2}$. Upper panels: free and excitonic electrons. Lower panels: trionic electrons and total dipole moment. (a) $N_h/N_e = 0.8$, (b) $N_h/N_e = 0.5$, and (c) $N_h/N_e = 0.2$.

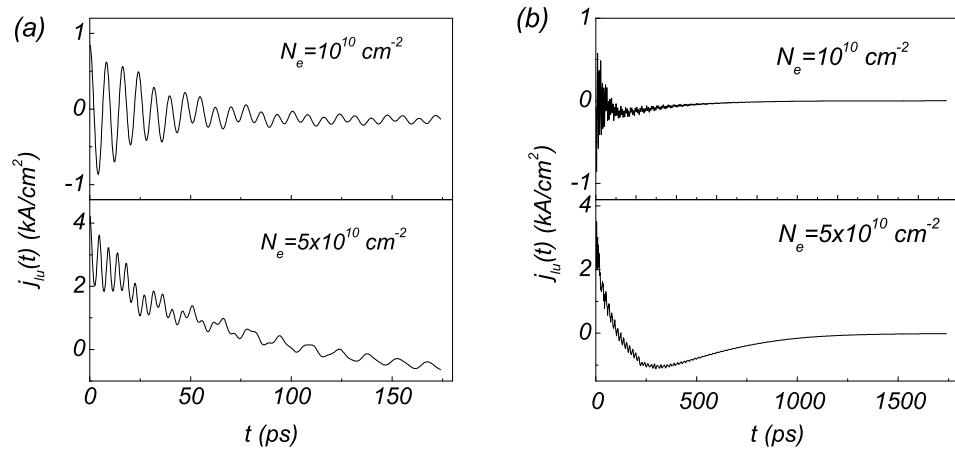


FIG. 6: Current density for $N_h/N_e = 0.8$. Upper panel: $N_e = 10^{10} \text{ cm}^{-2}$. Lower panel: $N_e = 5 \times 10^{10} \text{ cm}^{-2}$. (a) quantum beats for short times. (b) current relaxation for long times.

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- * Electronic address: paceitun@ull.es
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